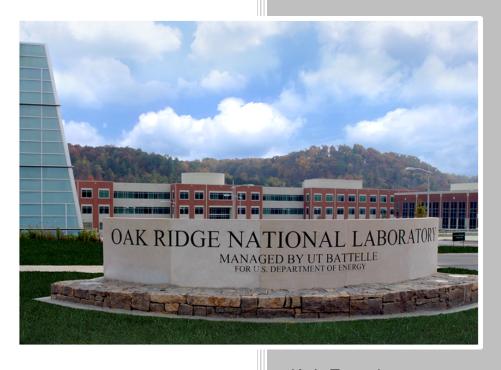
# Report on Design and Preliminary Data of Halden In-Pile Creep Rig



Approved for public release. Distribution is unlimited.

K. A. Terrani T. M. Karlsen Y. Yamamoto

September, 2015

#### **DOCUMENT AVAILABILITY**

Reports produced after January 1, 1996, are generally available free via US Department of Energy (DOE) SciTech Connect.

Website http://www.osti.gov/scitech/

Reports produced before January 1, 1996, may be purchased by members of the public from the following source:

National Technical Information Service 5285 Port Royal Road Springfield, VA 22161 *Telephone* 703-605-6000 (1-800-553-6847) *TDD* 703-487-4639 *Fax* 703-605-6900

**Fax** 703-605-6900 **E-mail** info@ntis.gov

Website http://www.ntis.gov/help/ordermethods.aspx

Reports are available to DOE employees, DOE contractors, Energy Technology Data Exchange representatives, and International Nuclear Information System representatives from the following source:

Office of Scientific and Technical Information PO Box 62
Oak Ridge, TN 37831 *Telephone* 865-576-8401 *Fax* 865-576-5728 *E-mail* reports@osti.gov

Website http://www.osti.gov/contact.html

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

#### ORNL/TM-2015/507

Under auspices of:
U.S. Department of Energy, Office of Nuclear Energy
Fuel Cycle Research and Development Program
Advanced Fuels Campaign

# REPORT ON DESIGN AND PRELIMINARY DATA OF HALDEN IN-PILE CREEP RIG

K. A. Terrani T. M. Karlsen Y. Yamamoto

September, 2015

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, TN 37831-6283
managed by
UT-BATTELLE, LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

# **CONTENTS**

	Page
CONTENTS	iii
LIST OF FIGURES	v
LIST OF TABLES	iv
1.INTRODUCTION	1
2.EXPERIMENT DESCRIPTION	2
2.1 TEST MATERIALS	3
3.RESULTS	
<ul><li>3.1 IN-PILE CREEP OF SiC</li><li>3.2 OUT-OF-PILE CREEP OF FeCrAl</li><li>3.3 IN-PILE CREEP OF FeCrAl</li></ul>	9 9
4.SUMMARY AND OUTLOOK	
REFERENCES	
Appendix A: Engineering Drawing of SiC Tensile Specimen for In-pile Creep Test	13
Appendix B: Engineering Drawing of FeCrAl Tensile Specimen for In-pile Creep Test	14

# LIST OF FIGURES

Figure		Page
Fig. 1.	Schematic of instrumented in-pile creep test rig.	2
Fig. 2.	Neutron flux spectra per unit lethargy in Halden IFA-744 test rig and HFIR PTP position (HFIR in blue)	4
Fig 3.	Neutron flux spectra in Halden IFA-744 test rig and HFIR PTP position.	5
Fig. 4.	dpa cross sections in barns for SiC, Fe, and Fe-13Cr-5Al as a function of neutron energy in MeV. The cross sections for Fe and FeCrAl are based on the NRT approach	5
Fig. 5.	Temperature, stress, and displacement as a function of neutron dose applied and experienced by SiC specimens.	7
Fig. 6.	Swelling and creep strain of SiC samples in the Halden experiment compared with swelling data from HFIR specimens from ref [16]	8
Fig. 7.	Instantaneous irradiation creep compliance of SiC measured in Halden at 300°C and compared with results from HFIR tests from ref. [7].	8
Fig. 8.	Out-of-pile creep strain as a function of time on FeCrAl alloys.	9
Fig. 9.	Temperature, stress, and displacement as a function of neutron dose applied and experienced by FeCrAl specimens.	10

# LIST OF TABLES

Table	Page
Table 1. Analyzed chemistry of the ORNL ATF FeCrAl alloys studied	3
Table 2. In-pile creep test parameters.	3
Table 3. dpa rate in SiC and Fe in HFIR and Halden calculated using various dpa cross sections.	6
Table 4. In-pile and out-of-pile creep strain rate experienced by FeCrAl specimens.	10

#### 1. INTRODUCTION

Given their exceptional oxidation resistance in high-temperature steam environments [1-2], SiC and FeCrAl alloys are leading candidates to replace Zr-based alloys in current light water reactors (LWRs) as accident tolerant fuel cladding. The goal of this study is to perform a sensitive examination of swelling and irradiation creep behavior of SiC and FeCrAl materials. These parameters are important material properties that dictate the stress and deformation behavior of the integral fuel pin. The viability of these cladding concepts to replace Zr-based alloys is contingent on their satisfactory performance under in-pile conditions as nuclear fuel cladding. The first steps to assess their viability, is to carry out detailed thermomechanical analysis of these structures as nuclear fuel cladding using fuel performance analysis tools. Therefore, it is essential to develop an accurate understanding of swelling and creep behavior of these materials through well-controlled experiments and use them as informed inputs into these fuel performance analysis tools.

Irradiation may result in swelling or enhance creep in materials among numerous other effects [3]. Irradiation swelling strictly refers to an isotropic volume increase in an unstressed material caused by the formation of irradiation-induce defects, resulting from the agglomeration of point defects and gas atoms inside the material. Irradiation Creep refers to a slow deformation at constant volume, in a material that is subjected to a stress below the yield stress; the role of irradiation is to accelerate creep mechanisms that occur outside irradiation. From an engineering perspective, the strict definition of irradiation creep refers to the observed difference in deformation from the material under stress with and without irradiation. The total deformation strain of the material under stress in the in-pile conditions is the sum of strains due to linear swelling, thermal creep, and irradiation creep, Eq. (1).

$$\varepsilon_{total} = \varepsilon_{swelling} + \varepsilon_{thermal\ creep} + \varepsilon_{irradiation\ creep}$$
 (1)

Swelling behavior of SiC has been studied extensively with comprehensive references available in the literature [4-5]. Thermal creep rate of SiC is insignificant at temperatures below 1400°C and can be ignored for LWR fuel cladding application [6]. Irradiation creep of SiC has received some attention and is undergoing continued investigation [7].

Swelling behavior of ferritic steels has received ample attention in the past and is known to be negligible in the dose range applicable to LWR fuel cladding [8]. Thermal creep is a strong function of alloy composition and microstructure and has received very limited attention in case of FeCrAl alloys in the temperature range of interest for LWR fuel cladding (300-400°C) [9]. Finally, irradiation creep has received some attention with rough estimates available for bcc metals in the literature [8].

#### 2. EXPERIMENT DESCRIPTION

This experiment involves in-pile creep testing of ATF FeCrAl and SiC specimens, with the aim of providing reliable in-pile data on creep compliance coefficient of these materials as a function of dose and temperature. Such data will be supportive of the qualification datapackage necessary for the eventual application of FeCrAl or SiC as a lead test rod (LTR) in a commercial LWR.

Only a limited temperature is probed that is directly applicable to the temperature of LWR fuel cladding under normal operating conditions. For FeCrAl alloys the test temperature was fixed at 350°C. In case of SiC, in order to exaggerate the swelling, the test temperature was kept constant at 300°C. The creep tests take advantage of in-pile instrumentation and control systems native to the Halden facility to collect accurate in-pile data with reliable environmental controls. Detailed description of this test rig is provided elsewhere [10], but briefly, they utilize adjustable mixture gas flow in annular regions of the capsule, coupled with multiple thermocouples in each capsule for precise temperature control along the length of the specimen. The stress on the creep specimens is controlled by direct gas pressure control inside a bellow attached to the creep specimen that is gripped at the other end. The creep strain is measured directly using a calibrated linear variable differential transformer (LVDT) that is in contact with the end of the specimen. Figure 1 shows a schematic of the in-pile creep test rig.

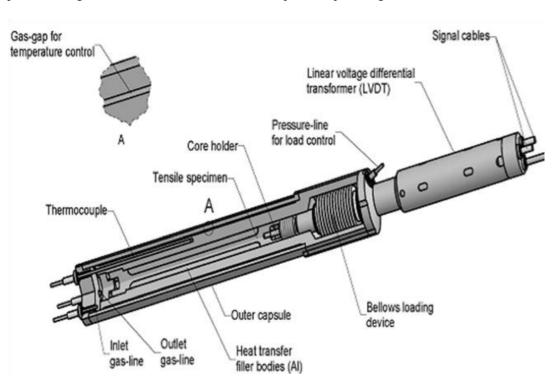


Fig. 1. Schematic of instrumented in-pile creep test rig from ref. [10].

#### 2.1 TEST MATERIALS

Two SiC specimens machined from high-purity chemical vapor deposited (CVD) SiC were examined in this study. The CVD variant, of purity >99.999%, was purchased from Dow Chemical Co. (Marlborough, MA). Also, three FeCrAl specimens from two distinct alloys were examined. The FeCrAl alloys were produced at ORNL and they were designated as C35M2 and C35MN5. The composition of the alloys is provided in Table 1. C35M2 alloy was hot-forged at 650°C with 75% area reduction (36mm diameter round bar to 16 mm square bar), followed by annealing at 650°C for 1h. Microstructure of this alloy consisted of deformed (elongated) grains. Note that the final annealing was applied to stabilize fine subgrain (SG) structure, with ~1-5 µm size inside of the elongated grains. C35MN5 alloy was hot-extruded at 800°C with 88% area reduction (2.9" diameter to 1.0" diameter), followed by annealing at 800°C for 1h. Microstructure consists of highly deformed (elongated) grains, which shows a dense dispersion of Nb-rich second-phase precipitates. Extensive detail on processing and properties of these alloys is provided elsewhere [11].

Analyzed composition [wt%] Heat Fe Cr Αl Mo Si Nb Y C N O S 79.67 13.06 0.010 0.004 0.0021 0.0017 0.0015 C35M2 5.15 1.97 0.12 C35MN5 78.68 13.02 5.08 1.99 0.21 0.97 0.032 0.003 0.0013 0.0028 0.0003

Table 1. Analyzed chemistry of the ORNL ATF FeCrAl alloys studied

#### 2.2 TEST CONDITIONS

Tensile specimens from the materials described were produced for this test. The exact drawings for the FeCrAl and SiC tensile specimens are provided at the end of this document as appendices. The specimens were tested in the stress-free and stressed conditions to determine the swelling-induced deformation only, as well as the swelling and creep-induced deformations combined. Note that a small stress (~5MPa) was applied under the stress-free conditions to ensure the specimen is in good contact with the LVDT. The test conditions are described in Table 2. Note that the stress on the SiC specimen was and remains maintained at 100 MPa. However the stress level on the FeCrAl specimens is varied over time to determine the creep rate as a function of stress. Also, since thermal creep may not be ignored for FeCrAl alloys, a separate series of out-of-pile creep tests at the exact temperature and stress levels were carried out at ORNL. In this manner the magnitude of thermal creep could be determined separately. The starting stress for FeCrAl alloys was set at a high value of 325 MPa that is just below the yield stress of these alloys at 350°C. This stress is representative of a value that is expected to develop in the thin cladding under fuel pin operation, particularly upon fuel-pellet contact.

Specimen Temperature [°C] Stress [MPa] C35MN5C-1 350 325 300 250 200 100 325 350 C35M2-1 350 325 300 250 200 100 325 350 C35M2-2 350 0 SiC-1 300 100 SiC-2 300

Table 2. In-pile creep test parameters.

#### 2.3 DPA RATE DURING IN-PILE TEST

Since irradiation creep is directly proportional to the damage induced in the material as a result of neutron fluence, it is essential to perform accurate calculation of damage production rate in these materials. This is particularly important when the results from the tests in a research reactor are to be compared with results from other research reactors or applied to predict the material behavior in commercial reactors. In order to perform dpa calculations, flux spectrum (flux per unit energy,  $\varphi_g[E]$ ) that is experienced by the specimen is necessary. The total flux,  $\phi$ , is the sum of the flux in each energy group. Flux spectra for HFIR (in the flux trap target (FTT) peripheral target position (PTP) at the midplane location) and Halden (IFA-744 rig at the location of creep capsules), are shown in Figure 2 and 3, where the former is normalized per unit lethargy. The total flux for the HFIR and Halden positions is  $4.46 \times 10^{15}$  and  $1.20 \times 10^{14}$  n/cm<sup>2</sup>-s, respectively. However, the spectra differ significantly, as shown in Fig. 2 where fast flux is roughly two orders of magnitude larger in HFIR.

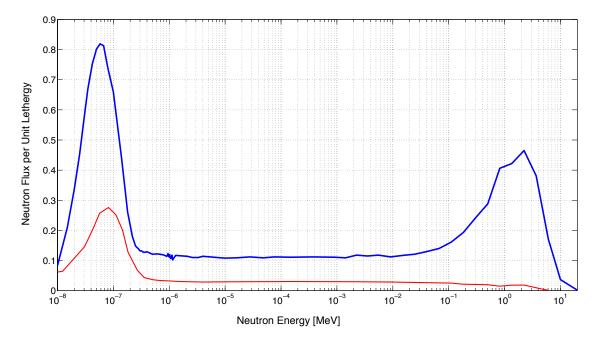


Fig. 2. Neutron flux spectra per unit lethargy in Halden IFA-744 test rig and HFIR PTP position (HFIR in blue).

The goal is to calculate the dpa (displacement per atom) rate in the materials of interest. The dap rate can be calculated as:

$$\dot{d} = \int \varphi_g(E)\sigma_d(E)dE \tag{2}$$

where  $\sigma_d$  is the displacement cross section. The displacement cross section for SiC [12] and pure Fe [13] are shown in Fig. 4. Also shown is the dpa cross section for Fe-13Cr-5Al alloy, calculated simply by weighing in the mole fraction of Cr and Al in the alloy and accounting for their dpa cross section from the same reference as for pure Fe. Note that the dpa cross section for the alloy was binned differently than that of pure Fe (larger energy bins) and appears smoother than the Fe dpa cross section.

The calculation in Eq. (2) was carried out using numerical methods and the dpa rate for these materials in Halden's IFA 744 and HFIR's FTT-PTP was estimated. The results are tabulated in Table 3. These results were also compared with the output from the SPECTER code [14] and they were in good agreement.

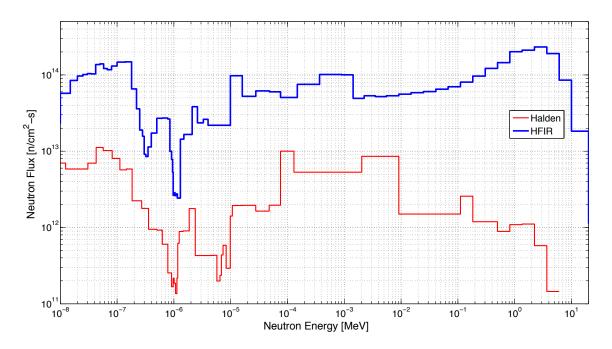


Fig 3. Neutron flux spectra in Halden IFA-744 test rig and HFIR PTP position.

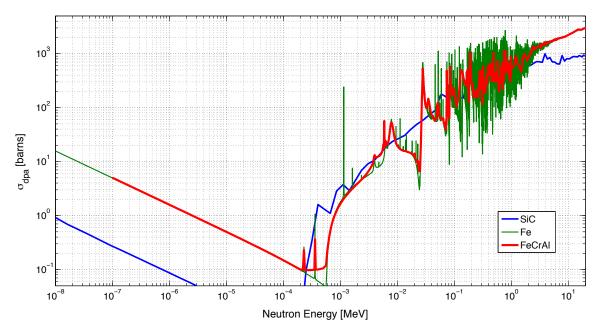


Fig. 4. dpa cross sections in barns for SiC, Fe, and Fe-13Cr-5Al as a function of neutron energy in MeV. The cross sections for Fe and FeCrAl are based on the NRT approach.

Table 3. dpa rate in SiC and Fe in HFIR and Halden calculated using various dpa cross sections

	dpa/s	dpa/HFIR <sup>1</sup> Cycle	dpa/month	dpa/1000h	dpa/EFPY
	CVI	D-SiC - dpa cross se	ction from [1	2]	
Halden IFA 744	3.59E-09		0.009	0.013	0.113
HFIR FTT-PTP	7.59E-07	1.57	1.97	2.73	23.84
Pure Fe – NRT <sup>2</sup>					
Halden IFA 744	4.94E-09		0.013	0.018	0.155
HFIR FTT-PTP	9.53E-07	1.98	2.47	3.43	29.94
FeCrAl – NRT <sup>2</sup>					
Halden IFA 744	4.63E-09		0.012	0.017	0.145
HFIR FTT-PTP	1.05E-06	2.18	2.72	3.78	32.99

<sup>&</sup>lt;sup>1</sup>HFIR cycle of 24 days is assumed. <sup>2</sup>NRT → Norget-Robinson-Torrens [15].

#### 3. RESULTS

#### 3.1 IN-PILE CREEP OF SiC

Figure 5 shows the results from in-pile measurement of displacement in SiC specimens along with the exact levels of applied stress and continuous measurement of temperature. The figure plots the high-quality displacement dataset as function of dose. A clear and significant departure in strain rate between the two samples is observed upon application of 100 MPa of stress on one of the specimens. Figure 6 compares the swelling and creep strain experienced by these specimens with discrete swelling strain data from specimens irradiated in HFIR where the two appear to be in good agreement.

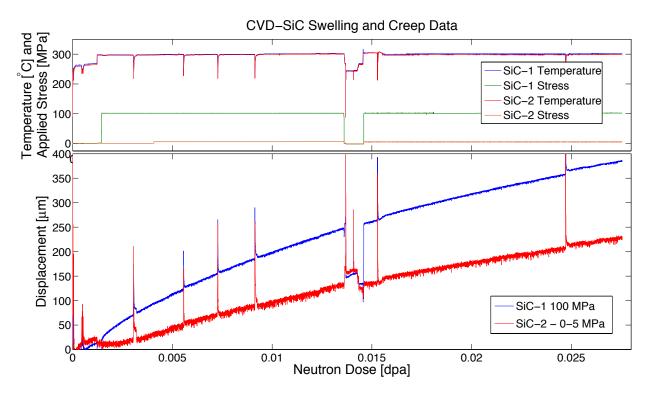


Fig. 5. Temperature, stress, and displacement as a function of neutron dose applied and experienced by SiC specimens.

As the described in Eq. (3), The difference in the instantaneous strain rate of stressed and stress-free SiC specimens, normalized per unit stress, is the instantaneous irradiation creep compliance of SiC.

$$\dot{\varepsilon}_{irradiation\,creep} = \frac{\dot{\varepsilon}_{stressed} - \dot{\varepsilon}_{swelling}}{\sigma} \tag{3}$$

The irradiation creep compliance is plotted as function of neutron dose in Fig. 7 and it appears to follow logarithmic creep behavior within this dose range. It is also compared with irradiation creep compliance data from bend stress relaxation (BSR) tests in HFIR [7]. The creep compliance measured here appears significantly larger than the prior HFIR results. The reason for this difference is not yet fully understood and may be partially due to the different grain boundary type and distribution between the two specimens used in these two separate tests. The large difference in the dose rate between these two irradiation experiments may also play an important role to result in this discrepancy.

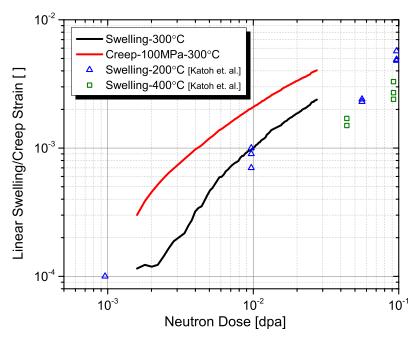


Fig. 6. Swelling and creep strain of SiC samples in the Halden experiment compared with swelling data from HFIR specimens from ref [16].

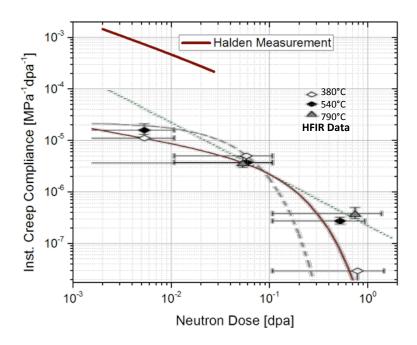


Fig. 7. Instantaneous irradiation creep compliance of SiC measured in Halden at  $300^{\circ}$ C and compared with results from HFIR tests from ref. [7].

#### 3.2 OUT-OF-PILE CREEP OF FeCrAl

Out-of-pile creep tests were performed on unirradiated FeCrAl specimens from the same exact batch of materials that was tested in Halden in irradiation creep capsules. The purpose of the out-of-pile tests was to separately determine the thermal creep rate in these alloys to then be able to distinguish all the distinct terms on the right-hand-side of Eq. (1). The tests were conducted at 350°C in an inert atmosphere on the C35M2 and C35MN5C alloys. The out-of-pile creep strain as a function of time is plotted in Figure 8. The thermal creep rate measured here for these alloys appears to be significantly (2-3 orders of magnitude) larger than the limited set of data reported in the literature for FeCrAl alloys at higher temperatures [9].

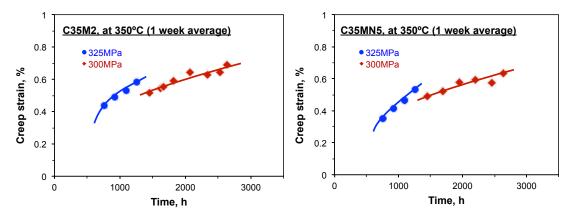


Fig. 8. Out-of-pile creep strain as a function of time on FeCrAl alloys.

#### 3.3 IN-PILE CREEP OF FeCrAl

Figure 9 shows the results from in-pile measurement of displacement in FeCrAl specimens along with the exact levels of applied stress and continuous measurement of temperature. The figure plots the displacement dataset as function of dose. A clear and significant departure in strain rate between the stressed and stress-free C35M samples is observed upon application of 325 MPa of stress on one of the specimens. In fact, within the dose range experienced by the C35M alloy, no swelling strain can be discerned in the stress-free sample. This is consistent with what is expected in ferritic alloys within this dose regime and up to at least a few tens of dpa (beyond what is expected within their lifetime as LWR fuel cladding) [8]. The displacement data for C35MN specimen appears unreliable and may hint at sensor failure during the test. Therefore, further analysis of the displacement data for this alloy is neglected.

Table 4 provides a comparison between the in-pile and out-of-pile creep strain rate experienced by these specimens. The out-of-pile and in-pile data are in very good agreement for the C35M alloy. This is indicative of the small relative magnitude of irradiation creep when compared with the thermal creep rate in this alloy. This is consistent with the estimates in the literature where irradiation creep in bcc metals is on the order of  $0.5 \times 10^{-6}$  MPa<sup>-1</sup>dpa<sup>-1</sup> [8].

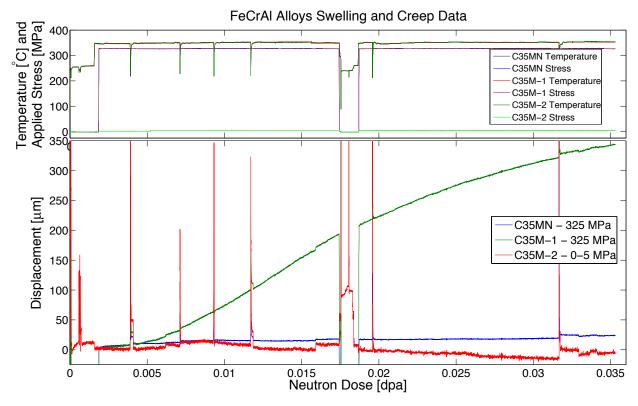


Fig. 9. Temperature, stress, and displacement as a function of neutron dose applied and experienced by FeCrAl specimens.

Table 4. In-pile and out-of-pile creep strain rate experienced by FeCrAl specimens

Alloy	Stress [MPa]	Dose [dpa]	Creep strain rate [h <sup>-1</sup> ]
	325	0	2.9×10 <sup>-6</sup>
	300	0	1.2×10 <sup>-6</sup>
C35M		0.007	2.4×10 <sup>-6</sup>
	325	0.012	3.7×10 <sup>-6</sup>
		0.030	1.6×10 <sup>-6</sup>
C35MN	325	0	3.4×10 <sup>-6</sup>
	300	0	1.0×10 <sup>-6</sup>

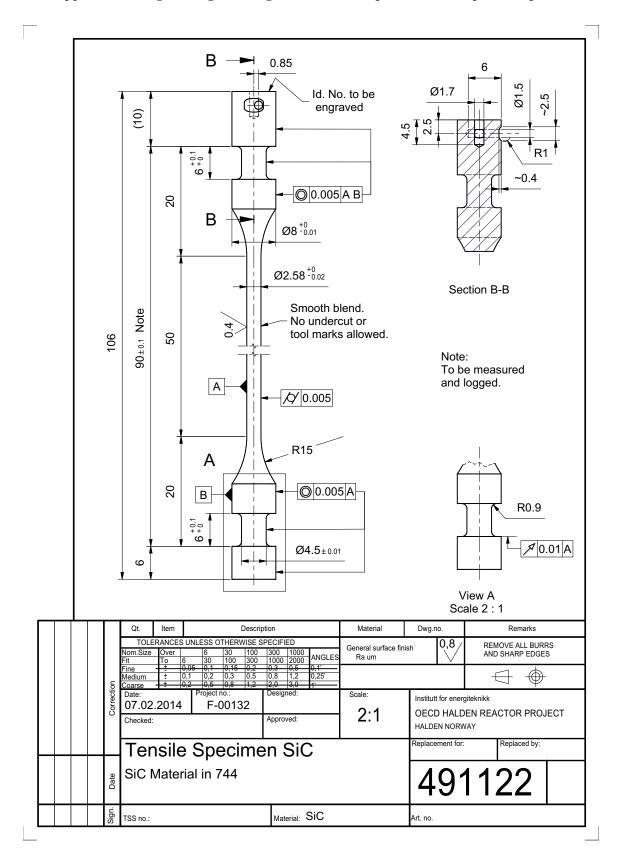
#### 4. SUMMARY AND OUTLOOK

A set of in-pile creep tests is ongoing in the Halden reactor on ORNL's candidate accident tolerant fuel cladding materials. These tests are meant to provide essential material property information that is needed for an informed analysis of these fuel concepts under normal operating conditions. These tests provide detailed information regarding swelling, thermal creep, and irradiation creep rates of these materials. The results to date have been compared with the limited set of information available in literature that is form irradiation tests in other reactors or out-of-pile tests. Most of the results are in good agreement with prior literature, except for irradiation creep rate of SiC. To elucidate the difference between the HFIR and Halden test results continued testing is necessary. The tests describe in this progress report are ongoing and will continue for at least another year.

#### REFERENCES

- [1] K. A. Terrani; B. A. Pint; C. M. Parish; C. M. Silva; L. L. Snead; Y. katoh, *Journal of American Ceramic Society* **2014**, *97* (8), 2331-2352.
- [2] B. A. Pint; K. A. Terrani; M. P. Brady; T. Cheng; J. R. Keiser, *Journal of Nuclear Materials* **2013**, 440, 420-427.
- [3] D. R. Olander, Fundamental Aspects Of Nuclear Reactor Fuel Elements. Technical Information Center: Springfield, VA, 1976; p 1-566.
- [4] Y. Katoh; K. Ozawa; C. Shih; T. Nozawa; R. J. Shinavski; A. Hasegawa, et al., *Journal of Nuclear Materials* **2014**, *448*, 448-476.
- [5] L. L. Snead; T. Nozawa; Y. Katoh; T. S. Byun; S. Kondo; D. A. Petti, *Journal of Nuclear Materials* **2007**, *371* (1-3), 329-377.
- [6] C. CARTER; R. DAVIS; J. BENTLEY, Journal of the American Ceramic Society 1984, 67 (11), 732-740.
- [7] Y. Katoh; L. L. Snead; C. M. Parish; T. Hinoki, Journal of Nuclear Materials 2013, 434 (1), 141-151.
- [8] F. A. Garner; M. B. Toloczko; B. H. Sencer, Journal of Nuclear Materials 2000, 276, 123-142.
- [9] S. Saunders; H. Evans; M. Li; D. Gohil; S. Osgerby, Oxidation of metals 1997, 48 (3-4), 189-200.
- [10] J. P. Foster; T. M. Karlsen *Update of Irradiation Creep and Irradiation Stress Relaxation of 316 and 304L Stainless Steel, HWR-1047*; OECD Halden Reactor Project: 2013.
- [11] Y. Yamamoto; B. A. Pint; K. A. Terrani; K. G. Field; Y. Yang; L. L. Snead, *Journal of Nuclear Materials* **2015**, *submitted*.
- [12] H. L. Heinisch; L. R. Greenwood; W. J. Weber; R. E. Williford, *Journal of nuclear materials* **2004**, 327 (2), 175-181.
- [13] A. Konobeyev; U. Fischer; L. Zanini In *Advanced evaluations of displacement and gas production cross sections for chromium, iron, and nickel up to 3 GeV incident particle energy*, Proceedings of the 10th International Topical Meeting on Nuclear Applications and Utilization of Accelerators, AccApp11. Knoxville, TN, US, 2011.
- [14] L. R. Greenwood; R. K. Smither SPECTER: Neutron Damage Calculations for Materials Irradiations, ANL/FPP/TM-197; Argonne National Laboratory: 1985.
- [15] M. J. Norgett; M. T. Robinson; I. M. Torrens, Nuclear Engineering and Design 1975, 33, 50-54.
- [16] Y. Katoh; T. Nozawa; L. L. Snead; K. Ozawa; H. Tanigawa, *Journal of Nuclear Materials* **2011**, 417 (1-3), 400-405.

Appendix A: Engineering Drawing of SiC Tensile Specimen for In-pile Creep Test



Appendix B: Engineering Drawing of FeCrAl Tensile Specimen for In-pile Creep Test

